Quantum dynamics in two and three-dimensional quasiperiodic tilings

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We investigate the properties of electronic states in two and three-dimensional quasiperiodic structures : the generalized Rauzy tilings. Exact diagonalizations, limited to clusters with a few thousands sites, suggest that eigenstates are critical and more extended at the band edges than at the band center. These trends are clearly confirmed when we compute the spreading of energy-filtered wavepackets, using a new algorithm which allows to treat systems of about one million sites. The present approach to quantum dynamics, which gives also access to the low frequency conductivity, opens new perspectives in the analyzis of two and three-dimensional models.

PACS numbers : 71.10.Fd, 71.23.An, 73.20.Jc

Since the discovery of quasicrystals [1], the nature of the eigenstates and of the energy spectrum, as well as the propagation of electrons in quasiperiodic potentials, have been the subject of many theoretical studies. One-dimensional ($1D$) quasiperiodic Hamiltonians have been widely investigated by means of numerical [2,3] and perturbative approaches [4,5]. Their spectrum consists in a set of zero width bands associated to critical eigenstates responsible of a sub-ballistic propagation of the electrons. In higher dimensions, the situation is more complex and, given the geometrical complexity of the structures, analytical treatments are difficult. Most of the studies have focussed on topologically trivial systems whose properties can be easily extracted from the $1D$ case (Fibonacci quasilattices [6,7], labyrinth tiling [8,9]). It has been shown that if the propagation is always sub-ballistic, the spectrum could be either absolutely continuous, singular continuous or any mixture. Other works based on exact diagonalizations, have also been performed on Penrose-like $2D$ and $3D$ systems. In $2D$, there is some evidence that eigenstates are critical [10] and responsible of an anomalous diffusion [11]. In $3D$ systems, the nature of eigenstates remains unclear [12]. In this context, it is of great interest to determine whether critical states are generic of *true* quasiperiodic systems especially in the $3D$ case, and to study the physical properties induced by such states.

In this letter we study in a tight-binding approach, the electronic properties of the of the $2D$ and $3D$ generalized Rauzy tilings (GRT) [13]. We discuss the nature of the eigenstates by computing their localization degree for small systems (~ 5500 sites). This analyzis suggests that the eigenstates are critical and rather more extended at the band edges than at the band center. To go beyond these exact diagonalizations, we introduce a powerful algorithm that allows us to study the quantum dynamics for very large systems (up to 10^6 sites here). Sub-ballistic propagation laws are clearly established and the energy dependence of the exponents confirms that states are more mobile (almost ballistic) close to the band edges than in the center. From this point of view, these results provides evidence of critical states in the $2D$ and $3D$ GRT. Finally, we discuss the low frequency conductivity which can be directly related to the quantum diffusion [14–16].

The GRT are codimension one quasiperiodic structures that can easily be built in any dimension D by the standard cut and project method [13]. Here, we focus on approximant structures that are obtained for rational cut directions. These tilings have a complex topological structure with sites of coordination number ranging from $D+1$ to $2D+1$. as displayed in Fig. 1 for $D=2$.

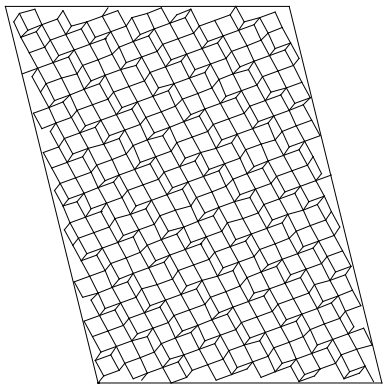

FIG. 1. A unit cell of the 10^{th} approximant of the $2D$ GRT with sites.

This geometrical complexity implies that they cannot be separated into a combination of $1D$ systems as in the Fibonacci quasilattices [6] or as in the labyrinth tiling [8]. We thus expect that their spectral and dynamical properties are representative of other $2D$ or $3D$ Penrose-like quasiperiodic structures.

We consider noninteracting electrons described by the following tight-binding Hamiltonian :

$$H = \sum_{\langle i,j \rangle} t \, |i\rangle\langle j|, \tag{1}$$

where $t = 1$ is the hopping energy between nearest neighbor sites, and where $|i\rangle$ denotes an orbital localized on the site i. As discussed in [13], the Hamiltonian matrix with periodic boundary conditions is straightforwardly written as a band (Toeplitz-like) matrix. This is due to a natural indexing of the sites known as the conumbering which classify them according to their local environment. The study of the spectrum of H has already been presented in Ref. [17], and we focus here on the nature of the eigenstates. Therefore, we have computed by exact numerical diagonalization, the participation number P of each normalized eigenstate $|\psi\rangle$ defined by : $P(\psi) = \left(\sum_i |\langle i|\psi\rangle|^4\right)^{-1}$. For a given system size, this quantity measures the localization degree of the state $|\psi\rangle$ considered. We display in Fig. (2), for the $2D$ and $3D$ GRT, the participation ratio $p = P/N$ as a function of the energy (N is the number of sites per unit cell). Qualitatively, we observe in both systems that, despite strong fluctuations, the most extended eigenstates are globally located near the spectrum edges and the more localized ones in the central part.

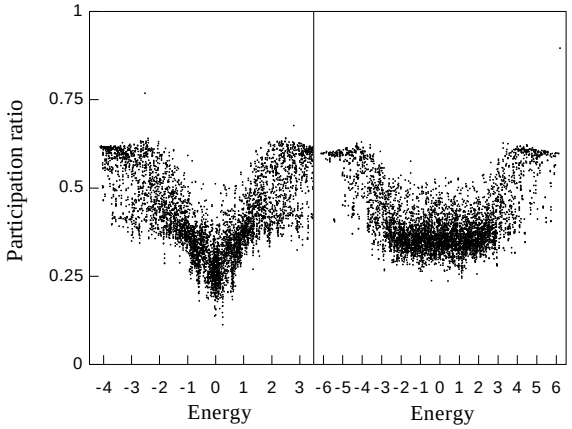

FIG. 2. Participation ratio as a function of the energy for the 14^{th} approximant (5768 sites) of the $2D$ GRT (left) and for the 13^{th} approximant (5536 sites) of the $3D$ GRT (right).

Another interesting information is given by the scaling of the mean participation number (averaged over the whole spectrum) versus the number of sites N. Indeed, when the system size increases, the mean participation number converges towards a finite value if the states are exponentially localized and goes to infinity otherwise. As shown in Fig. 3, this mean participation number behaves as N^γ with $\gamma_{2D} \simeq 0.965 \pm 0.001$ and $\gamma_{3D} = 0.975 \pm 0.003$, indicating a weakly critical behaviour of the eigenstates. Note that this is only an average behaviour that conceals strong fluctuations from one energy to another as it has already been shown in Penrose-like tilings [12].

In the $2D$ GRT, the exponent is greater than the one obtained for the octagonal tiling [18] (codimension 2) where $\gamma = 0.87 \pm 0.05$. This difference can be understood by invoking the codimension of the structures that gives an indication on the geometrical complexity in terms of local environment. Intuitively, we indeed expect low codimension structures to be weakly *quasiperiodic*, and to have weakly *critical* states. The recent level statistics analyzis done in the $2D$ GRT [19] also corroborates this assumption. Finally, up to the numerical precision, we have $\gamma_{2D} < \gamma_{3D}$. It means that for a fixed codimension (here 1), the localization of the eigenstates increases when the dimension is lowered, as it is the case in disordered systems. To confirm the trends observed with diagonalizations, we have studied the spreading of energy-filtered states on much larger systems of about one million sites.

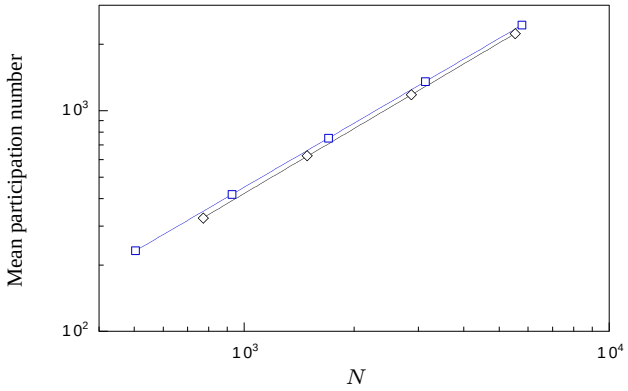

FIG. 3. Mean participation number as a function of the number of sites for the $2D$ (□) and the $3D$ (◇) GRT.

The diffusion of wave packets can be measured by the mean square spreading :

$$X^2(E,t) = \langle (\hat{X}(t) - \hat{X}(0))^2 \rangle_E \tag{2}$$
$$= \langle A^\dagger(t) A(t) \rangle_E, \tag{3}$$

where $A(t) = [\hat{X}, \mathrm{e}^{-\mathrm{i}Ht}]$. $\hat{X}(t)$ is the Heisenberg representation of the position operator along the x direction and $\langle \ldots \rangle_E$ denotes the average value over the eigenstates of energy E. We stress that $X^2(E,t)$ evolves with time even though it is a mean value over eigenstates.

$X^2(E,t)$ is related to $\sigma(E_{\mathrm{F}}, \omega)$ the conductivity at frequency ω and Fermi energy E_{F} [16]. In the limit of small frequencies one has :

$$\langle \sigma(E_{\mathrm{F}}, \omega) \rangle_\Delta = e^2 \int_0^\infty \mathrm{e}^{\mathrm{i}\omega t} \left\langle n(E_{\mathrm{F}}) \frac{\mathrm{d}^2 X^2(E_{\mathrm{F}}, t)}{\mathrm{d}t^2} \right\rangle_\Delta \mathrm{d}t \tag{4}$$

where $n(E_{\mathrm{F}})$ is the density of states at the Fermi energy, e is the electron charge, and $\langle \ldots \rangle_\Delta$ means an av-

erage over a range Δ of values of $E_{\rm F}$. Note that at zero frequency, the integral in (4) is equal to the diffusivity $D(E_{\rm F}) = \lim_{t\to\infty} {\rm d}X^2(E_{\rm F},t)/{\rm d}t$ and one recovers the Einstein formula $\sigma(E_{\rm F},0) = e^2 n(E_{\rm F}) D(E_{\rm F})$.

To investigate the quantum diffusion, we consider an approximant of the GRT with a sufficiently large unit cell so that the eigenstates are Bloch waves characterized by a wave vector $\mathbf{k}$. As for the exact diagonalizations we restrict our study to the $\mathbf{k} = 0$ subspace, i.e. to the eigenstates which are invariant by translation from one unit cell to another. Note that for an approximant the operator $A(t)$ is invariant by translation of a unit cell and thus lets invariant the subspace of $\mathbf{k} = 0$ states. We calculate $X_0^2(E,t)$ defined by :

$$X_0^2(E,t) = \langle A^\dagger(t) A(t) \rangle_{E,\mathbf{k}=0} \tag{5}$$

$$X_0^2(E,t) = \frac{{\rm Tr}\left\{A^\dagger(t)\delta(E-H)A(t)\right\}}{{\rm Tr}\left\{\delta(E-H)\right\}} \tag{6}$$

Actually, $X_0^2(E,t)$ is a good approximation of $X^2(E,t)$ provided that t is lower than the time required to significantly couple orbitals separated by a distance $l_x/2$ during the time evolution. In this study, we have always considered wave packet evolutions for such times. The operator trace Tr is restricted to the subspace $\mathbf{k} = 0$. An efficient trick to evaluate the traces is to use states $|\psi\rangle$ having a constant presence probability on each site $|\langle i|\psi\rangle|^2 = 1/N$ and a random phase on each orbital of the unit cell. We thus approximate ${\rm Tr}\{B\}$ by $N\langle\psi|B|\psi\rangle$. In fact, we should perform an average over several random phases configurations but we have checked that, thanks to the great number of sites in the unit cell, we only need to consider one random phase state $|\psi\rangle$ to get a good estimate of the traces. Moreover, to eliminate fluctuations, we have performed a convolution of the $\delta(E-H)$ by a Gaussian operator of typical width $\Delta = W/100$ where W is the total bandwidth [20]. This is equivalent to compute the spreading of energy filtered wavepackets. These parts of the calculation are rather standard and in fact the crucial step is to evaluate $A(t)|\psi\rangle$.

We start from the expansion of the evolution operator ${\rm e}^{-{\rm i}Ht}$ using a basis of Chebyshev polynomial $P_i(E)$ of the first kind. These polynomials are associated to the weight $\rho(E) = 1/\left(\pi\sqrt{4b^2-(E-a)^2}\right)$. This expansion converges provided the spectrum of H (computed here using the standard recursion method [21]) is included in the interval $[a-2b, a+2b]$ [20]. The decomposition of the evolution operator is written as :

$${\rm e}^{-{\rm i}Ht} = \sum_{i=0}^{\infty} c_i(t) P_i(H), \tag{7}$$

with $c_p(t) = {\rm i}^p h_p J_p(-2bt)$ where $h_0 = 1$, $h_{p>0} = 1/2$, and where $J_p(x)$ is the Bessel function of order p [22,23]. At large p and fixed x, one has : $J_p(x) \simeq 1/\sqrt{2\pi p}\,(x\,{\rm e}/2p)^p$ so that the sum in Eq. (7) converges quickly and can be truncated. The polynomials P_i obey the following relation :

$$bP_{i+1}(H) = (H-a)P_i(H) - bP_{i-1}(H), \tag{8}$$

with $P_0(H) = 1$ and $P_1(H) = (H-a)/2b$. Then, introducing $|\phi_i\rangle = P_i(H)\,|\psi\rangle$ and $|\psi_i\rangle = [\hat{X}, P_i(H)]\,|\psi\rangle$ and using Eqs. (7) and (8), one readily obtains :

$$b|\phi_{i+1}\rangle = (H-a)|\phi_i\rangle - b|\phi_{i-1}\rangle \tag{9}$$

$$b|\psi_{i+1}\rangle = (H-a)|\psi_i\rangle - b|\psi_{i-1}\rangle + [\hat{X}, H]|\phi_i\rangle. \tag{10}$$

Solving this latter system allows one to compute efficiently :

$$A(t)|\psi\rangle = \sum_{i=0}^{\infty} c_i(t)|\psi_i\rangle, \tag{11}$$

and thus $X_0^2(E,t)$ with an arbitrary accuracy given by the order of the truncation.

All the results discussed here have been obtained in the 22^{nd} approximant (755476 sites) for the $2D$ GRT and in the 21^{st} approximant (1055026 sites) for the $3D$ GRT. We have computed, for both tilings and for several energy-filtered wave packets, the time dependence of $R_0^2(E,t) = X_0^2(E,t) + Y_0^2(E,t) + Z_0^2(E,t)$ where x, y and z are three orthogonal directions ($Z_0 = 0$ in $2D$).

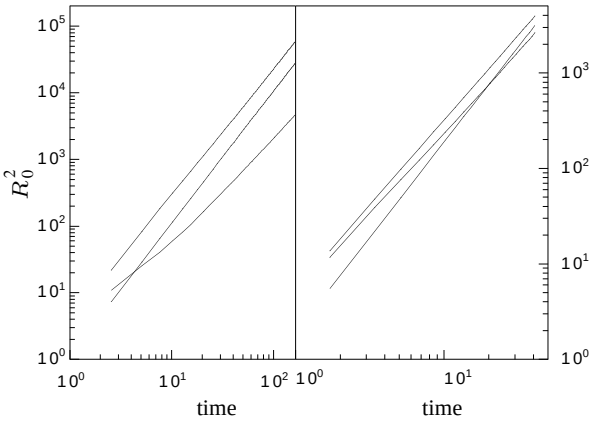

FIG. 4. Behaviour of $R_0^2(E,t)$ for three energy-filtered wave packets in the $2D$ GRT (left) and in the $3D$ GRT (right).

As displayed in Fig. 4, one has $R_0^2(E,t) \sim t^{2\beta(E)}$ at large times with $0 < \beta(E) \leq 1$. The observation of such a sub-ballistic propagation for large approximant is consistent with the existence of critical eigenstates. The energy dependence of the diffusion exponent β shown in Fig. 5 also clearly confirms the tendency observed with exact diagonalizations since the diffusion exponent is, in both systems ($2D$ and $3D$), larger at the band edges than at the spectrum center. In the $3D$ GRT, we have even obtained an almost ballistic propagation ($\beta \simeq 1$). In addition, the shape of the density of states is nearly

free- electron like at band edges which is also consistent with the less localized eigenstates at these energies. If this general trend is similar to what is observed in the octagonal tiling [11] and in the Penrose tiling [12], it is the opposite of what occurs in the $3D$ icosahedral Amman-Kramer tiling [12]. Note also that in disordered systems, states at band edges are more localized than in the band center. The parameters that determine the nature of band edges states are obviously not yet understood.

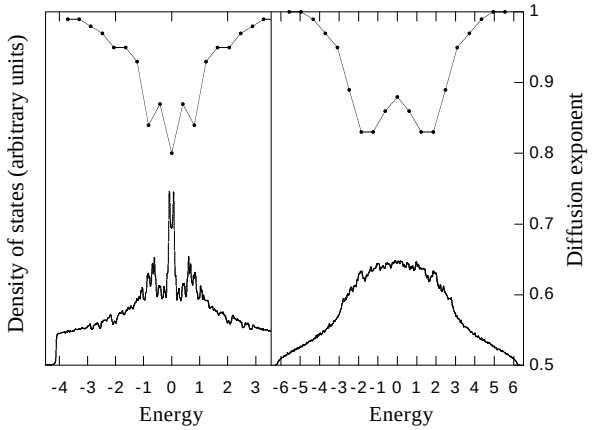

FIG. 5. Normalized density of states (lower curve) and diffusion exponents (upper curve) of the $2D$ GRT (left) and of the $3D$ GRT (right).

We would like to emphasize that the scaling laws are valid for $R_0^2(E,t)$ varying by $3-4$ orders of magnitude in $2D$ and $2-3$ orders of magnitude in $3D$. In both cases the number of sites N contained in a volume $R_0^2(E,t)$ in $2D$ and $R_0^3(E,t)$ in $3D$ varies by $3-4$ orders of magnitude. This is much better than with exact diagonalization for which the typical variation of N is 1 order of magnitude.

The power-law behaviour obtained for $R_0^2(E,t)$ allow us to explicitely calculate the low frequency conductivity. Indeed, assuming $X^2(E_{\rm F},t) \simeq C(E_{\rm F})t^{2\beta(E_{\rm F})}$, one obtains, using Eq. (4) :

$$\sigma(E_{\rm F},\omega) \simeq e^2 n(E_{\rm F})\, C(E_{\rm F})\, \Gamma\left(2\beta+1\right) (\mathrm{i}/\omega)^{2\beta-1}, \quad (12)$$

where Γ is the Euler Gamma function. For the GRT, the diffusion exponent $\beta(E_{\rm F})$ is always greater than $1/2$ (superdiffusive propagation) so that although the system is non periodic, its dc conductivity $\sigma(E_{\rm F},\omega=0)$ is infinite for all $E_{\rm F}$. Note that experiments are qualitatively explained if $\beta < 1/2$ [16] but such values of β probably require a strong quasiperiodic potential.

To conclude, we would like to point that the calculation of quantum diffusion presented here, implies a computational load proportionnal to the number N of sites of the system whereas exact diagonalizations implies a load which is of order N^3 or $N^2 \ln N$ for the best algorithms. This is the main reason of the great efficiency of the present method and of its ability to treat large systems. Note that in the context of large scale electronic structure calculations, order N algorithms raise much interest and can also be much more efficient than exact diagonalizations [24]. As a consequence, this approach should open new perspectives in the investigation of $2D$ and $3D$ models.

We thank Claude Aslangul for a careful reading of the manuscript.

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